Wide single-mode tuning of a 3.0–3.8-μm, 700-mW, continuous-wave Nd:YAG-pumped optical parametric oscillator based on periodically poled lithium niobate

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A new optical parametric oscillator (OPO) for the mid-infrared wavelength region of 3–3.8 μm with an idler output power of up to 1.5 W has been developed. The singly resonant OPO is pumped by a single-mode, 10-W, continuous-wave Nd:YAG laser and consists of a bow-tie ring cavity with a fan-out periodically poled lithium niobate crystal and a low-finesse intracavity air-spaced etalon. The single-frequency idler output can be continuously tuned over 24 GHz with 700-mW power by tuning of the pump laser. The tuning was demonstrated by recording of an absorption line of ethane with photoacoustic spectroscopy. © 2002 Optical Society of America

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For spectroscopic applications in the mid infrared (wavelength region 2.5–5 μm), continuous-wave (cw) optical parametric oscillators (OPOs) based on quasi-phase-matched materials such as periodically poled lithium niobate (PPLN) can be very useful. These OPOs can cover a wide operating range and usually have a reasonable output power of several hundred milliwatts.[1,2] For direct absorption techniques, such as cavity-ringdown spectroscopy, low powers in the milliwatt region are sufficient.[3] However, for a number of spectroscopic applications high power combined with continuous single-frequency tuning of the OPO is essential. In many cases, such as molecular beam spectroscopy (see, e.g., Ref. 4) and photacoustic spectroscopy,[5,6] high powers improve the performance of the applied technique.

A typical scanning range should cover at least one but preferably multiple rovibrational transitions of the species under investigation. For molecular spectroscopy this demands an OPO linewidth of less than 10 MHz over a tuning range of tens of gigahertz for the mid-infrared wavelength region.

Bosenberg *et al.* first showed high-power operation of a cw, singly resonant optical parametric oscillator (SRO) based on PPLN in 1996.[1] They demonstrated more than 1 W of tunable cw idler output power in the wavelength range 3.3–3.9 μm. However, because of the pump laser used (a single-transverse-mode, multiple-axial, diode-pumped 1.064-μm Nd:YAG laser, a Lightwave Electronic Corporation prototype), they could not reach a linewidth of better than 2.2 GHz for the idler.

SROs have a high oscillation threshold and therefore need high pump powers. Doubly resonant OPOs have much lower oscillation thresholds and can therefore be pumped by low-power single-frequency (diode) lasers. An additional advantage of diode pumping is the wide tunability of the pump wavelength, which one can use to tune the idler beam of the OPO. A recent example is a 20-MHz-linewidth doubly resonant OPO with 18-mW idler output covering the 2.2–3.7-μm region.[7] Potentially, with its pump source this laser could obtain over 100 GHz of continuous tuning, but the doubly resonant cavity design limits its tunability to 10 GHz.

A SRO directly pumped by a diode laser was reported by Klein *et al.*,[8] and continuous tuning with a diode-pumped SRO system was also demonstrated by this group.[9] In the latter study, their pump system was a master oscillator–power amplifier with 4-MHz linewidth, tunable over 60 GHz. With this design they were able to demonstrate 56-GHz continuous tuning of the idler wave. However, this system covered the 2.01–2.19-μm wavelength region, which is much easier to operate in than the widely used 3-μm idler region. Furthermore, this system has a small operating range and a lower idler output power of 200 mW compared with other SRO systems.

In this study we aim for a high-power (>1 W), single-frequency idler output SRO with a wide operating range (3.0–3.8 μm), a large continuous pump-tuning range (tens of gigahertz), and a small linewidth. The continuous pump-tuning range of 24 GHz that is demonstrated in this study has been surpassed only by Klein *et al.*,[9] and to our knowledge it is the highest continuous tuning range ever reported with a Nd:YAG-pumped SRO system. The tuning of this Nd:YAG source makes our system unique.

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We achieve this tuning by pumping the OPO with a cw single-longitudinal-mode master oscillator–power amplifier system (Lightwave M6000). In this system the master oscillator (MO) is a state of the art nonplanar ring oscillator that produces frequency-stable, narrow-linewidth, single-axial-mode output at a Nd:YAG wavelength of 1064 nm. The MO is continuously tuned over 24 GHz by changes in the temperature of the nonplanar ring oscillator crystal. The power amplifier uses 160 W of diode pump power to amplify the output of the master oscillator while contributing nearly undetectable changes in noise or beam quality. The complete system produces an output power of up to 15 W, with narrow linewidth (<5 kHz over 1 ms) and low noise, which can be frequency tuned over 24 GHz.

When the pump power is changed, the laser beam characteristics (i.e., divergence, pointing) change significantly. For this reason a combination of a half-wave plate (New Focus, Santa Clara, Calif.) and a polarizing beam splitter (OFR, Caldwell, N.J.) are used to change laser power without affecting the beam characteristics (Fig. 1). The basic setup of the cavity design of the OPO is similar to the one used earlier.[2,10] A 10-cm lens focuses a 10-W pump beam into the PPLN crystal. The PPLN crystal (Crystal Technology, Palo Alto, Calif.), which is 5 cm long and 0.5 mm thick, has a fan-out grating design as described earlier,[2] with periodicities ranging from 29.3 to 30.1 μm. To prevent photorefractive damage, we keep the crystal at 180 °C in a commercially available oven (Super Optronics, Gardena, Calif.), and the crystal is temperature stabilized at 0.1 °C. The input and output faces of the PPLN are antireflection coated for 1.064 μm, 1.5–1.7 μm, and 3.0–3.8 μm. The PPLN is placed in a four-mirror ring cavity in which the signal beam at 1.5–1.7 μm is resonating (free spectral range cavity, 400 MHz). In this SRO the two curved mirrors (radius of curvature, 10 cm; VLOC, New Port Richey, Fla.) and the two flat mirrors (QTF, Oldsmar, Fla.) have high reflectivity R for the signal wave ($R > 99.9\%$ at 1.5–1.7 μm) but high transmission T for the pump and idler waves ($T > 90\%$ and $R < 0.25\%$ at 1064 nm and $T > 80\%$ at 3.0–3.8 μm).

The combination of a single-mode pump source and a single-mode ring cavity resonating at the signal wavelength produces a nonresonant but single-frequency idler beam with tuning characteristics that are similar to those of the pump laser.

The idler output and pump depletion are shown in Fig. 2. The oscillation threshold was found to be 3 W for the pump, whereas the pump depletion was as great as 70% for 9-W pump power (maximum idler power, 1.5 W). The pump depletion was highly dependent on proper focusing of the pump beam into the PPLN crystal. A 10-cm lens focused the pump beam to a waist of 100 μm in the center of the crystal. The pump depletion is still low compared with that of other SRO systems; a possible reason for this is that the mirrors are not reflective enough. Rotating the pump polarization and the PPLN crystal orientation by 90° might improve the performance in future.

With this setup the OPO cavity works fairly stably, with infrequent mode hops. Even though it is not absolutely necessary, single-mode operation is enhanced by an intracavity etalon. This enhancement will also allow cavity mode-hop tuning in the future.[2] We used either an uncoated 0.4-mm-thick solid-state YAG etalon (VLOC) or an air-spaced etalon with a free spectral range of ~150 GHz and a reflectivity of 10% (Laser Optik, Garbsen, Germany). We carried out the first experiments with a solid-state YAG etalon to minimize the losses. The output power of the OPO with and without this etalon was similar, but unfortunately the etalon was not selective enough to prevent all mode hops in the OPO cavity. In addition, mode-hop tuning the cavity with this etalon was very limited because of walk-off losses when the solid-state etalon was rotated.

Instead of the solid-state etalon we decided to use a 10%-reflectivity, air-spaced etalon. Even though

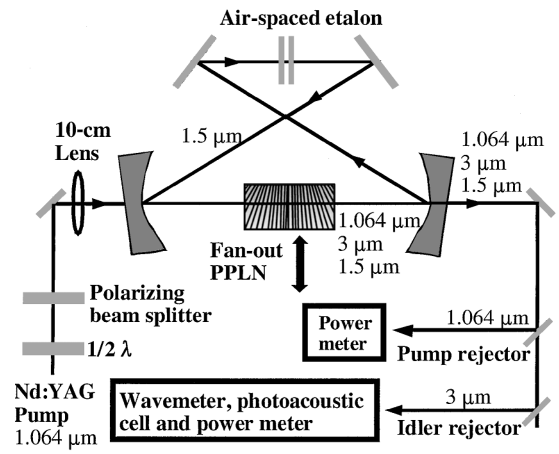

Fig. 1. Experimental setup of the cw SRO. The pump beam is focused into the 50-mm-long fan-out PPLN with a 10-cm lens. A combination of a half-wave plate and a polarizing beam splitter is used to change the pump power. The cavity is resonant for the signal beam at 1.5–1.7 μm and consists of two flat mirrors and two curved mirrors with 10-cm radii of curvature. The idler wavelength at 3.0–3.8 μm is sent to a photoacoustic cell and a wavemeter. An intracavity air-spaced etalon is used to enhance frequency.

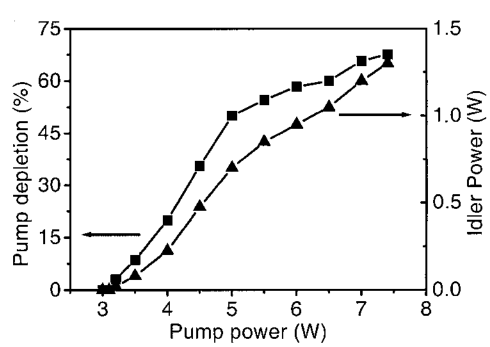

Fig. 2. Idler output and pump depletion versus pump input power for the SRO operating at an idler wavelength of 3.3 μm. The oscillation threshold is 3.0 W, and a maximum idler power of 1.5 W is observed with a pump power of 9 W.

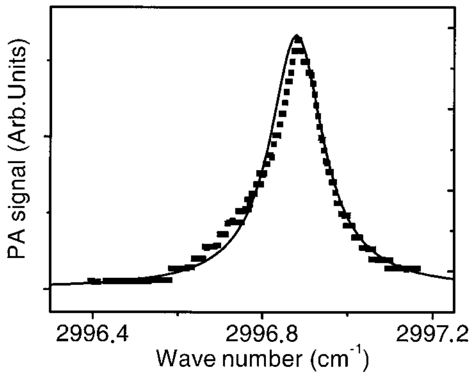

Fig. 3. Single-frequency tuning of the pressure-broadened (0.75-atm) $\nu 7$, $v = 1 \leftarrow 0$, $K = 4 \leftarrow 3$, Q-branch transition ($J = 4-39$) of ethane is demonstrated by use of photoacoustic (PA) spectroscopy. The experimental results are shown by the scatter graph, and the spectrum of ethane from the Hitran database is shown by the solid curve.

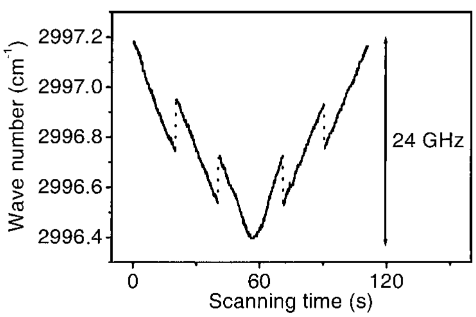

Fig. 4. While the idler wavelength was monitored with a wavemeter, the pump source of the OPO was tuned by a change in its driving voltage. The whole range of the pump laser was scanned, and after ~60 s it was scanned back. The mode hops in the idler wavelength (dashed lines) are caused by mode hops in the pump source. A total tuning range of 24 GHz is found. The resolution of the picture was limited by the resolution of the wavemeter and the data acquisition card of the computer.

this etalon gave more losses, the advantages of better OPO stability and a longer mode-hop tuning range were found to be more important. Walk-off losses were minimized by placement of the air-spaced etalon between the two flat mirrors in the second focal point of the SRO. Inserting the air-spaced etalon caused the output power to decrease from 1.5 W to ~700 mW.

Single-frequency tuning was demonstrated by recording of the pressure-broadened (0.75-atm) $\nu 7$, $v = 1 \leftarrow 0, K = 4 \leftarrow 3$, Q-branch transition ($J = 4-39$) of ethane by use of photoacoustic spectroscopy.[6] Figure 3 shows the absorption line of 10 parts in 10^6 of ethane in nitrogen, compared with the same absorption line in the Hitran database. The idler wavelength (Fig. 4) was monitored by an infrared wavemeter (Burleigh WA1000 IR, Fishers, N.Y.). The total idler tuning was found to be 24 GHz, with mode hops after 12 GHz of tuning. The two mode hops observed in the idler wavelength are caused not by mode hops in the SRO but by mode hops in the master oscillator of the pump laser. These hops in the pump source cause no trouble, because they are predictable and because the pump laser always hops back compared with the direction of tuning (Fig. 4). This hopping back means that some parts of the scan are done twice, but no part of the scan is skipped. If the mode hops of the pump laser could be eliminated, then truly continuous tuning over 24 GHz could be achieved. From the noise in the measured absorption line we estimate the frequency stability of our system to be better than 50 MHz. In the near future we will demonstrate this by using a 300-MHz high-finesse etalon; we expect a stability of better than 1 MHz.

Present research is concentrated on further improvements of the OPO setup. Optimizing the reflectivity and the FSR of the air-spaced etalon should improve the long-term stability of the SRO. By rotation of the polarization of our pump source and the PPLN crystal orientation by 90°, a better pump depletion with a higher output power is expected. Furthermore, by combining our pump tuning with mode-hop tuning,[2] we expect that the total continuous tuning range will be increased considerably. With these improvements this OPO will certainly be a very good source for most spectroscopic applications. We will apply this system in life science by combining it with photoacoustic spectroscopy[6] to develop a sensitive trace-gas detector. Ethane, which was measured in this study, is emitted from biological samples when cell walls' membranes are damaged and is therefore an important gas in biological studies.[11] We expect the lowest detectable concentration of ethane in air to be at sub-part-in-10^9 (parts per billion) levels.

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